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A CONTINUOUS CORROSIVITY MONITORING DEVICE FOR NAVAL ENVIRONMEN--ETC(U)
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A CONTINUOUS CORROSIVITY MONITORING DEVICE
FOR NAVAL ENVIRONMENTS

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12 November 1980

PROGRESS REPORT
AIRTASK NO. R0220101
Work Unit No. DG202

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Prepared for
NAVAL AIR SYSTEMS COMMAND
Department of the Navy
Washington, D.C. 20361

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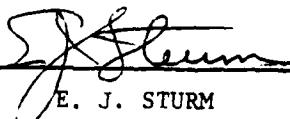
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER 14 NADC-80198-60	2. GOVT ACCESSION NO. AD-A095728	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) A Continuous Corrosivity Monitoring Device for Naval Environments.		5. TYPE OF REPORT & PERIOD COVERED Progress
7. AUTHOR(s) Vinod S. Agarwala		6. PERFORMING ORG. REPORT NUMBER
9. PERFORMING ORGANIZATION NAME AND ADDRESS Aircraft and Crew Systems Technology Directorate Naval Air Development Center Warminster, Pennsylvania 18974		8. CONTRACT OR GRANT NUMBER(s)
11. CONTROLLING OFFICE NAME AND ADDRESS Naval Air Systems Command Navy Department Washington, DC 20361		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS AIRTASK R0220101 Work Unit No. DG202
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		12. REPORT DATE 12 November 1980
		13. NUMBER OF PAGES 26
		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION, DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release, distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Corrosion monitoring probe, galvanic cell, environmental corrosivity, relative humidity, digital recording system, corrosion current transients, salt-SO ₂ environment, aircraft carrier environment		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Based on the principles of an electrochemical galvanic cell, a probe, with related data recording system, has been adopted for continuous monitor- ing of the corrosivity of naval environments. The probe has been used successfully to monitor the corrosivity on an aircraft carrier during an eight-month deployment. Current transients recorded during this period were analyzed and related to similar measurements made in controlled laboratory environments containing high humidity, chloride or chloride/SO ₂ .		

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TABLE OF CONTENTS

	<u>Page No.</u>
LIST OF TABLES	ii
LIST OF FIGURES.	ii
INTRODUCTION	1
EXPERIMENTAL DETAILS	2
PROBE DESIGN AND INSTRUMENTATION.	2
ELECTROCHEMICAL CONCEPTS.	3
PROCEDURES.	4
RESULTS AND DISCUSSION	5
CONCLUSIONS.	7
FUTURE PLANS	8
ACKNOWLEDGEMENTS	9
REFERENCES	10

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L I S T O F T A B L E S

<u>TABLE</u>		<u>PAGE NO.</u>
I	Effects of Environmental Variables on Probe Output	11
II	Open-Circuit Potentials ($E_{\text{corr.}}$) and Corrosion Rates	12
	($I_{\text{corr.}}$) after 24 Hours in 3.5% NaCl Solution (Immersed)	12

L I S T O F F I G U R E S

<u>FIGURE</u>		<u>PAGE NO.</u>
1	Environmental Corrosivity Monitoring Probe and the Related Instrumentation Schematic	13
2	Galvanic Current Transients of Copper/Steel Probe Exposed to Various Laboratory Simulated Environments	14
3	Galvanic Current Transients of Copper/Aluminum Probe Exposed to Various Laboratory Simulated Environments	15
4	Galvanic Current Transients of Steel/Aluminum Probe Exposed to Various Laboratory Simulated Environments	16
5	Galvanic Current Transients of Steel/Aluminum Probe Exposed to an Aircraft Carrier Environment	17

I N T R O D U C T I O N

Currently the assessment of corrosivity of an environment is made on the projected figures obtained on a cumulative basis from outdoor, longterm exposure tests. Tests such as salt fog, total or alternate immersion, are primarily designed from such projections to produce an accelerated laboratory test environment. Although these test methods have been quite successful in evaluating various materials, they are too often indiscriminately used. In situations where rapid weather changes control the nature of the environment or the locations are inaccessible, a true evaluation of the corrosivity of the environment is not possible. Any extrapolation of the results obtained from laboratory tests may be inaccurate at best, and deceptive, at worst.

The corrosion problems of naval aircraft are of a very severe and varied nature. In particular, sulfur from the carrier stack gases combined with sea spray provides a uniquely hostile environment (reference (a)). The aggressive naval aircraft carrier and marine environments not only undermine the structural integrity of the aircraft but also affect the navigational and communication systems as well. Inaccessible and critical functional areas of aircraft are very sensitive to even the slightest corrosion damage. An assessment of corrosion problems in such systems is almost impossible through the conventional corrosion tests. Corrosion of avionics, gear and gear housings, bearings and wing flaps etc. are well documented failures. In addition, the present trend to miniaturization and large scale integration of computer hardware in aircraft will even further increase their susceptibility to corrosion. Therefore, it is important that an assessment of the actual corrosivity of the environment be made before any laboratory testing procedure can be developed to simulate the environment.

In the present work a probe has been developed using well known techniques, (references (b)-(g)) which can monitor the corrosivity of the environment on a continuous basis and also can be placed in areas which are otherwise inaccessible. This probe is a series of electrochemical galvanic cells comprised of plates of two different metals such as aluminum and steel or steel and copper sandwiched together alternately separated by an electrical insulator and encapsulated in epoxy. When short circuited through an external electronic circuit of a zero-resistance ammeter, the condensed moisture and the pollutants in the environment create an electrolyte (thin liquid film) on the surface of these plates which completes the circuit and develops a galvanic potential between the two. The magnitude of this cell current gives a measure of the corrosivity of the condensed film (the environment). The concepts of this technique were originally investigated by Sereda (reference (b)) who used platinum and zinc as the galvanic couple to measure the time of wetness during atmospheric exposure. Sereda's time of wetness measuring technique was arbitrarily based on a potential of 0.2V (a voltage drop across a 10M Ω resistor) as a timer was started when the potential drop exceeded 0.2V. Sereda chose this as a reference voltage developed by the Pt/Zn couple when exposed to an environment which contained 85% R.H. Tomashov and co-workers (reference (c)) independently utilized a similar approach and developed closely spaced galvanic cells for monitoring time of wetness of panels exposed to a high humidity environment. Later Kucera and Mattsson

(reference (d)) modified this technique and the principles used earlier by Sereda and Tomashov to develop galvanic cells of copper and steel and suggested that they can be used to monitor atmospheric corrosion rates. Lauer and Mansfeld (reference (f)) and Mansfeld and Kenkel reference (g) further explored Kucera and Mattsson's suggestions and designed several galvanic couples made up from copper, steel, aluminum and zinc metals. More recently, Mansfeld (reference (b)) made a study of the effect of weather conditions and correlated the output of the couples with the corrosion rate. In the present work a study has been made to develop a wider use of this technique for specifically measuring the severity of the marine weather and the pollutants of the naval environment on corrosion behavior of steels and aluminum alloys.

EXPERIMENTAL DETAILS

PROBE DESIGN AND INSTRUMENTATION

The design of the probe was based on the approach used by earlier investigators (references (e)-(g)). Three galvanic couples, copper-steel, copper-aluminum and steel-aluminum were made from approximately 1 mm thick sheet metal stock of commercial purity. "Virgin" teflon of 0.75 mm thick was used as insulators. Ten pieces (5 x 2.5 x 0.1 cms) of each of the two dissimilar metals (one as a cathode and the other as an anode) were sandwiched together alternately and insulated by the teflon spacers. Two holes were drilled through this assembly for inserting nylon bolts to hold it together. All similar metal pieces (e.g., copper to copper or steel to steel) were short-circuited by thin electrical conductors and ended with two terminals, one for the cathode (e.g., copper) and the other for the anode (e.g., steel). The whole array of these plates was then thinly coated with stop off lacquer on the sides and potted in epoxy resin leaving only one long edge surface and the two wire terminals exposed. The potted assemblies were then sufficiently cured at room temperature before the exposed edge of the probe was surface ground and polished to a 600 grit finish. The total exposed area of the probe was approximately 5 cm² for each metal.

The galvanic cell current output from the probe was recorded on a periodic basis (short intervals) using the electronic devices such as a zero resistance ammeter (Z.R.A.), an analog to digital converter; i.e., a digital panel meter (DPM) of very high impedance and a data logger. The schematic of the set up is shown in Figure 1. The ZRA was put together by using a potentiostat (Aardvark Model PEC-1) in which the voltage between the working (W) and reference (R) terminals was set to zero. A standard resistance box was connected between the reference (R) and counter (C) terminals for converting the galvanic current, I_g , into a potential drop for amplification and recording purposes. The digital data logging system used was a commercially available instrument from Precision Digital (Model 1040). The data logger was programmed to record probe output every 10 minutes during exposure. The choice of digital recording instruments eliminated the use of a log converter in this study as current variations of up to three orders of magnitude could be conveniently recorded without changing the resistance between (R) and (C). A 10K Ω resistor was adequate to give a current range of 1 μ A to 1mA in naval environments.

ELECTROCHEMICAL CONCEPTS

The principles involved in the use of this corrosivity monitoring probe are those of corrosion of galvanically coupled metals. The driving force in the galvanic corrosion is the difference in electrode potentials between two dissimilar metals where the more active metal becomes the anode and the other the cathode. Usually the cathode or cathodic metal corrodes very little or not at all and corrosion of the anode or anodic metal is increased significantly. In other words, both metals are polarized so that each corrodes at a new rate. The extent of this polarization potential, η , of both the anodic and cathodic metals determines the magnitude of galvanic current flowing between two dissimilar metals and is expressed by the Stern-Geary equation

$$I_g = I_{\text{corr}} \left[(10)^{\frac{\Delta\eta}{\beta_a}} - (10)^{-\frac{\Delta\eta}{\beta_c}} \right] \quad (1)$$

After a series expansion and neglecting the higher terms assuming $\frac{\Delta\eta}{\beta_a}$ and $\frac{\Delta\eta}{\beta_c}$ being small, the equation (1) can be written as

$$I_g = I_{\text{corr}} \times 2.3\Delta\eta \left[\frac{1}{\beta_c} + \frac{1}{\beta_a} \right] \quad (2)$$

where β_c and β_a are the Tafel slopes of the cathodic and anodic polarization respectively (reference (i)). However, when applied to corrosion reactions in which the cathodic (reduction) reaction is under diffusion control (i.e., at the limiting current) and the IR drop is significant as one would expect in the atmospheric corrosion phenomena, the situation is equivalent to a large or infinite value of β_c in equation (2).

Hence, the equation (2) can be simplified as

$$I_g = I_{\text{corr}} \times 2.3 \left(\frac{\Delta\eta}{\beta_a} \right) \quad (3)$$

But, for a particular couple at some steady state, the polarization potential $\Delta\eta$ becomes a constant. Hence

$$I_g = \text{Constant} \times I_{\text{corr}} \quad (4)$$

i.e., the galvanic current, I_g becomes the corrosion current of the anode. This relationship is also in agreement with Mansfeld and Kenkel (reference g)). However, it is important to note that the corrosion current, I_{corr} , does depend upon the electrochemical nature of the cathodic metal as it alters the value of $\Delta\eta$ in equation (3). In this analysis it has been assumed that the cathodic metal behaves more like an oxygen depolarizer therefore it undergoes the least corrosion. The extent by which the polarization potential, $\Delta\eta$, shifts is primarily dependent upon three important environmental variables, temperature, relative humidity and corrosivity (i.e., nature of the environment such as presence of O_2 , salts, acids and other gaseous and particulate matter). In other words, the output of a galvanic probe will be dependent upon these parameters. Experimentally the observed changes will be directly related to the corrosive nature of the environment.

The details of the mechanism of galvanic corrosion phenomena are described elsewhere (references (c), (g), (i), (j), and (k)). Probably the best explanation of how a galvanic couple behaves in a corroding system is offered by the mixed potential theory (reference (j)).

PROCEDURES

The experimental program to explore the applicability of the corrosion monitoring probes utilized two laboratory environments and one natural environment to achieve three different exposure conditions: (1) a continuous high humidity (near 100% R.H.) environment; (2) an accelerated laboratory corrosion test environment; and (3) an aircraft carrier. The galvanic probes used to study these environments were copper/steel, copper/aluminum and steel/aluminum. Both copper and aluminum were commercially pure metals and the steel used was an AISI 1040 grade. Use of the three different galvanic probes for testing was made to determine the effect of potential difference as the driving force on the galvanic current developed in various media and to determine the performance of one metal over another in so far as the corrosion of anodic metal was concerned.

Near 100% relative humidity in the test chamber was created by passing compressed air through two vessels containing deionized water one after another and by using gas bubblers (fritted glass tubes). The humidity and temperature of the chamber were monitored by a digital humidity and temperature measurement system manufactured by Thunder Scientific Corporation (Model HS-1CHDT-2R). In experiments where chloride and acid were introduced into the chamber, the air bubbling vessels contained 1% NaCl and 6M HCl solutions respectively, instead of deionized water. With this procedure approximately 0.2 gm/m^3 of NaCl and 4 gms/m^3 of HCl could be present at any time in the chamber. The galvanic current transient curves (dotted) shown in Figures 2-4 represent the results of these test conditions.

In the accelerated laboratory corrosion testing, a salt fog cabinet meeting the requirements of ASTM B117-73, Appendix I, was used. A 5% NaCl solution was sprayed into the chamber as a continuous mist to simulate nearly wet conditions as experienced on naval aircraft carriers. In the tests where SO_2 laden salt fog was required, SO_2 gas was introduced in the cabinet for one

hour four times a day while salt fog was continuously sprayed. The details of this testing are described elsewhere (reference (1)). The probe exposure data obtained by this accelerated laboratory corrosion testing are shown as continuous curves of galvanic current transients in Figures 2-4.

In the aircraft carrier exposure tests, the probe was installed on a radar tower about 20 feet above the flight deck of the aircraft carrier, USS JOHN F. KENNEDY (reference (m)). The galvanic current measuring instruments were located in the radar room to isolate them from the flight deck. The sensitivity of the recording system was set low so that at high relative humidity (>85%), the output of the probe was low (1-2 microamps). This way the full scale range setting on the recording system could measure up to 500 microamps. The probe output was recorded for a period of eight months, from June 1978 to March 1979, during which the carrier was deployed to the Mediterranean. At the end of the mission, the current transient records were analyzed and condensed as shown in Figure 5. The probe used in this study was a steel/aluminum couple.

RESULTS AND DISCUSSION

The galvanic current-exposure time output for the three probes, copper/steel, copper/aluminum and steel/aluminum, evaluated here in various corrosive environments are shown in Figures 2, 3 and 4, respectively. It has been shown that after an initial surface conditioning of the probes which may take 5-10 hours, the probe output tends to become steady and depending upon the environment present in the chamber, the galvanic currents take an orderly relative position on the scale of corrosivity. Generally, it was found that as the corrosivity of the environment was increased the galvanic current measured by the probes also increased in the same order. This order of corrosivity was independent of the metals used in the probe but differed in magnitude depending on the couple. A comparative evaluation of their performance in various environments has been summarized in Table I. On comparison it was noted that the steel/aluminum probe was a little less sensitive to environmental variables. However, when the environment was of a highly acidic nature as in the case of 100% R.H + HCl (cf. Table I and Figure 4), the steel/aluminum probe showed an order of magnitude increase when compared to copper/steel. The copper/aluminum probe also showed a correspondingly higher output in the acid environment. Here, the logical explanation lies in the behavior of aluminum which becomes very active at very low pH and high chloride ion concentrations. This was also confirmed by the fact that almost the same corrosion potentials were measured for these couples (copper/aluminum and steel/aluminum) when immersed in a 3.5% NaCl solution (cf. Table II). A review of the electrochemical data obtained for three metal systems shown in Table II also indicated that as compared to the corrosion potentials of the couples, the open circuit potentials of the uncoupled anodic metals do not change significantly. It can therefore be concluded that in galvanic coupling the cathodic metal is mostly polarized and the galvanic current measured is largely due to the corrosion of the anodic member of the couple only; concurrent results were also reported by Walker (reference (n)). Although these results refer to an immersed state, it is believed that similar electrochemical polarization occurs on thin electrolyte

films (condensed environment on the probe surface) as well, except for some kinetic effects which originate due to concentration limiting diffusion parameters. An earlier study by the author (reference (o)) relates to these conclusions.

Based on the results obtained by the corrosivity monitoring probes as summarized in Table I, the environments can be arranged as follows in the order of increasing corrosivity:

Moisture < (Moisture + Cl) < (Moisture + SO₂)
 < Salt Spray < (Salt Spray + SO₂) < (Salt Spray + SO₂ + Soak)

The increase in corrosivity from one environment to the next was almost an order of magnitude with the exception of the one which contained HCl. Some parallel tests on panels exposed to these environments also exhibited similar behavior and confirmed the results as shown by the probe output.

The condensed data of the aircraft carrier exposure test (reference (m)) after replotting, as shown in Figure 5, exhibit several periods of high corrosion activity (currents). In the first 40 days the steel/aluminum probe showed very little corrosion activity (less than 5µA) indicative of normal marine weather and similar to those obtained from the probe in 5% salt spray environment (cf. Table I). Between 40 to 160 days there appeared to be periods of wetness resembling the data obtained in salt spray and SO₂ test environment. However, in the last 100 days of deployment considerable corrosive activity was recorded with galvanic currents as high as 500µA at some instance. This correlates with the reports from carrier personnel that the weather in the first four months of deployment was generally mild, whereas that of the last four months was characterized by storms and high seas. It also correlates with corrosion rates exhibited by the aluminum alloys exposed on the carrier when inspected at four months and eight months. Exfoliation attack was slight at four months and severe after eight months exposure (reference (m)). High current peaks were indicative of extremely corrosive conditions such as those created in the laboratory by periods of high acidity, chloride, SO₂ and almost wet conditions. In addition, the deposition of particulate matter such as heavy metals, soot and grease on the surface of the probe may have further accelerated the corrosion effects significantly. A qualitative analysis (AES) of the smut deposited on the probe showed detectable amounts of Cu, Ni, Pd, Cr, Fe and Mn. It has been reported by LaQue (reference (p)) that the presence of heavy metals like copper in sea water can accelerate corrosion of aluminum significantly. The results shown by the copper/aluminum galvanic probe in the laboratory accelerated test (cf. Table I) seem to substantiate this conclusion.

The results obtained by the probe in the carrier exposure tests were indicative of the diversities of environment where the weather in question changed frequently from very wet and corrosive to almost normal or calm.

It then raises a question whether the corrosivity monitoring probe was actually responding to such changes in the carrier environment. To affirm that, the galvanic probes were subjected to a cyclic accelerated environmental test. It was comprised of a 30 minute spray of synthetic sea water (ASTM D1141-75, Section 6) in the spray cabinet followed by a 30 minute flow of SO_2 gas (flow rate of 1 cc/min/ft³ of cabinet volume) and then all activity stopped for the next two hours (called the soak period). At the end of the soak period the cycle starts all over again and continues, repeating every three hours. The probe exposure data obtained from these tests are shown in Figure 6. The I_g vs. time curves demonstrate a good correlation with the changing nature (corrosivity) of the environment in the cabinet. The gap in the arrows in Figure 6 indicates that the peak corrosivity of the environment was repeated after approximately three hours which coincided well with the cyclic period regulated in the cabinet. The variations in the time of occurrence of these peaks were probably the result of initial surface stabilization of the probe and the frequency with which the cabinet cover was open. In subsequent long exposure tests these variables were better regulated. It was noted (cf. Figure 6) that the steel/aluminum probe was not as sensitive to the environmental changes as the other two. In order to determine the behavior of this probe on a long term basis, this test was continued for a period of five days. The results obtained from the copper/steel probe were even more interesting. As shown in Figure 7, the I_g vs. exposure time curve is made up of maxima and minima of the corrosion currents which were consistently repeated after every three hours and became very reproducible as the exposure continued. The ascending part of the curve coincided with the time when SO_2 gas was flowing into the cabinet after salt spray had ceased. Thus, the peaks correspond to the point in time where the probe experienced the highest SO_2 concentration; i.e., maximum acidity and maximum wetness as the salt fog continues to remain in the chamber for at least 1/2 hour after its flow has stopped. The extrapolated dotted lines in the curve (cf. Figure 7) were drawn to make the corrections in the curve, because during those periods, the cabinet cover was opened for examination which probably allowed the environment to escape (change) and therefore affected the results. The steady state values of the maxima and minima varied from 100 to 300 μA showing extremely high corrosive environmental changes. In Table I a comparative summary of the performance of various probes used in the cyclic test has been given. From the actual panel exposure testing it was observed that this synthetic sea salt + SO_2 + soak cycle environment was probably the most corrosive of all accelerated laboratory corrosion testing environments (cf. Table I) and the type of attack produced had the closest resemblance to that obtained in the carrier environment.

CONCLUSIONS

Based on the principles of galvanic action a corrosion monitoring probe has been developed and evaluated for its usefulness in measuring the severity of the environment quantitatively. A comparative study of three galvanic probes, copper/steel, copper/aluminum and steel/aluminum showed that the copper/steel probe was most responsive to most varieties of environments in terms of its corrosion sensitivity and reproducibility when used on a continuous basis.

In a simulated accelerated cyclic corrosion test environment, the corrosion monitoring ability of the probe was found to be in agreement with the cyclic corrosive nature of the environment. The aircraft carrier exposure results obtained by the probe indicated a very high corrosion activity caused by the presence of high wetness, sea salt, SO₂ and deposition of particulate matter (most probably heavy metals).

FUTURE PLANS

Plans are underway to install a probe on the naval research ship, USS VANGUARD, which will be cruising from an east coast port. It will be possible to check frequently the meteorological records and correlate them with the probe current measurements which was difficult on an operational carrier.

The probe will also be installed in an electronic bay of a P-3 based at the Naval Air Development Center. In the electronic bays alternate condensation and evaporation of moisture take place. This will provide an opportunity to see if the probe is sensitive enough to quantitatively characterize on an aircraft a cyclic environment such as that.

A C K N O W L E D G E M E N T S

The author gratefully acknowledges the generous financial support of the Independent Research Board of the Naval Air Development Center, the Naval Air Systems Command, and the Commander, Naval Air Atlantic Fleet for the opportunity to perform the aircraft carrier exposure tests. The author is thankful to Mr. K. Briegel for experimental work and Dr. J. J. De Luccia and Mrs. S. J. Ketcham for their helpful suggestions.

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TABLE I. EFFECTS OF ENVIRONMENTAL VARIABLES ON PROBE OUTPUT

<u>Environment</u>	Probe Output, I_g , ^a μA		
	<u>Copper/ Steel</u>	<u>Copper/ Aluminum</u>	<u>Steel/ Aluminum</u>
100% R.H.	0.01	0.01	<0.01
100% R.H. + Cl^- ^b	0.1	0.5	0.01
100% R.H. + HCl ^c	28	260	220
100% R.H. + SO_2	1.5	3	0.5
5% NaCl Spray	7.5	8	1.5
5% NaCl Spray + SO_2	140	80	7
Syn. Sea Water + SO_2 + Soak (Cyclic Test)	120-300	80-200	15-30

^a I_g values at steady-state after 20 hours of initial exposure.

^b Cl^- in the test chamber carried by the air when purged through 1% NaCl solution.

^c HCl in the test chamber carried by the air when purged through 6M HCl solution.

TABLE II. OPEN-CIRCUIT POTENTIALS ($E_{\text{corr.}}$) AND CORROSION RATES ($I_{\text{corr.}}$) AFTER 24 HOURS IN 3.5% NaCl SOLUTION (IMMERSED).

<u>Material</u>	$E_{\text{corr.}}^I$, V (Vs SCE)	<u>Galvanic Couple</u>	$E_{\text{corr.}}^{II}$, V Vs SCE	$I_{\text{corr.}}^a$ $\mu\text{A}/\text{cm}^2$
Aluminum (C.P.)	-0.730	Steel/Al	-0.740	12
AISI 1040 Steel	-0.610	Cu/Al	-0.720	30
Copper (C.P.)	-0.240	Cu/Steel	-0.680	20

a Corrosion rates refer to the anodic member of the couples only.

$E_{\text{corr.}}^I$ and $E_{\text{corr.}}^{II}$ refer to potentials of uncoupled anodes and cathodes, and galvanic couples after one hour immersion respectively.

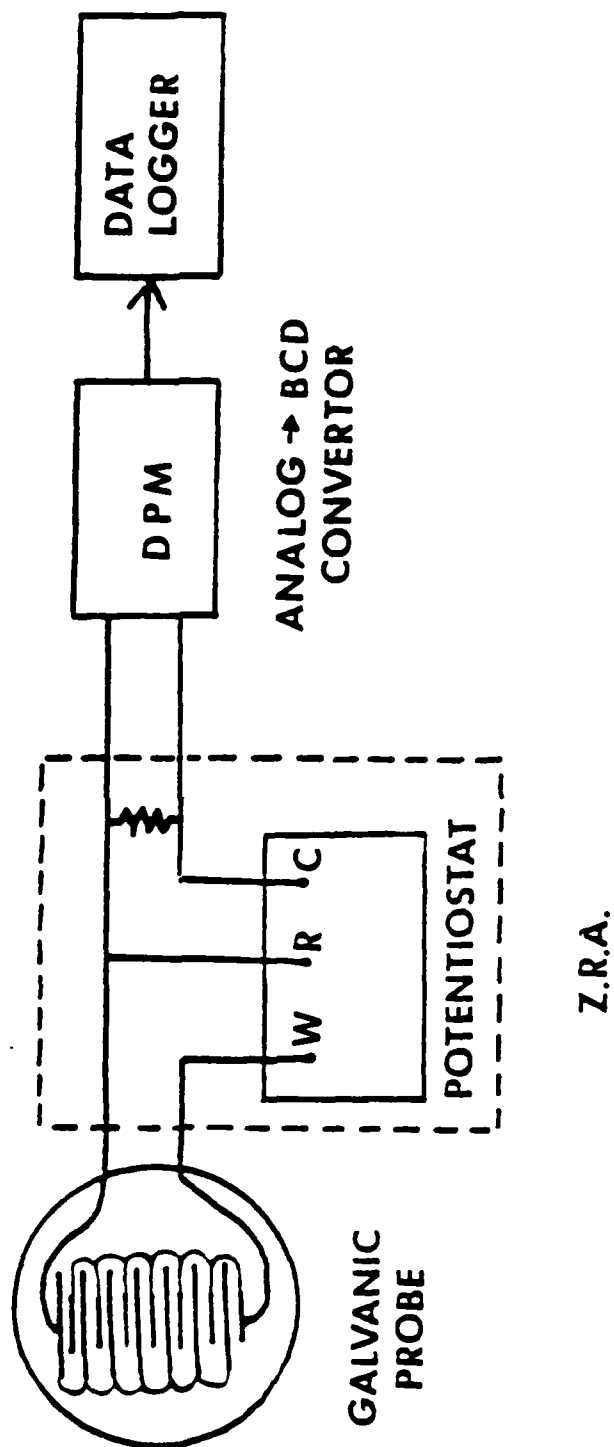


FIGURE 1. ENVIRONMENTAL CORROSIVITY MONITORING PROBE AND THE RELATED INSTRUMENTATION SCHEMATIC.

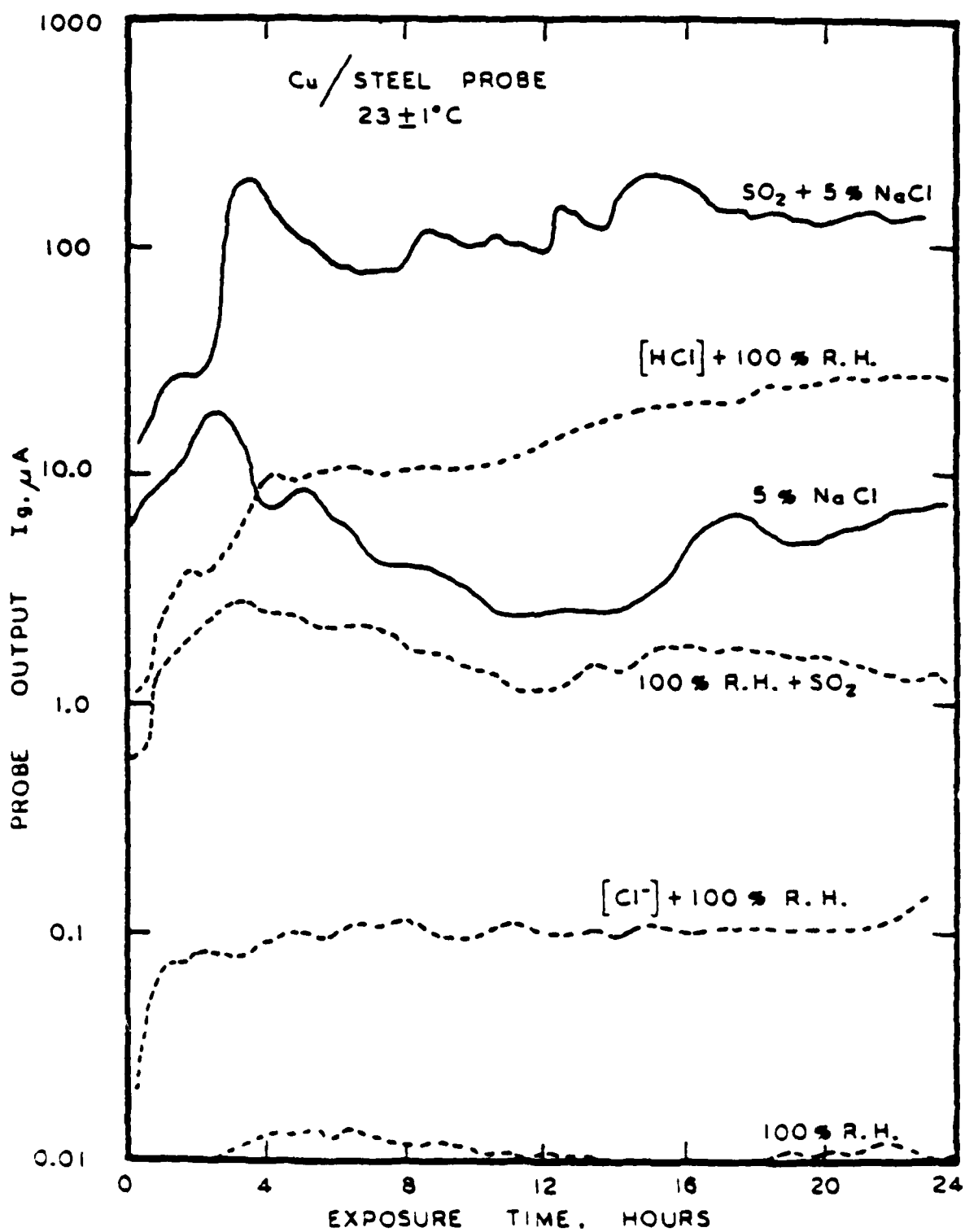


FIGURE 2. GALVANIC CURRENT TRANSIENTS OF COPPER/STEEL PROBE EXPOSED TO VARIOUS LABORATORY SIMULATED ENVIRONMENTS.

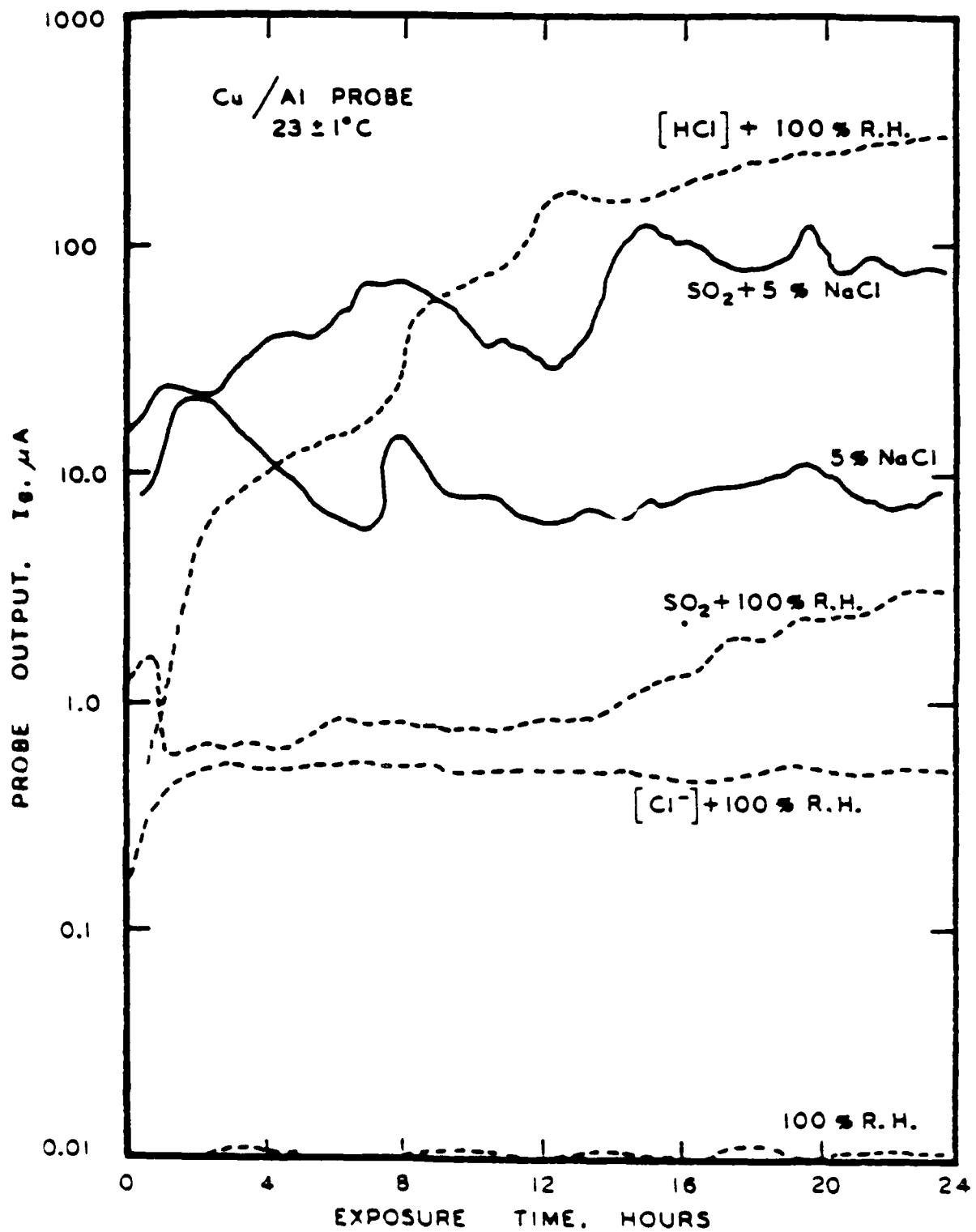


FIGURE 3. GALVANIC CURRENT TRANSIENTS OF COPPER/ALUMINUM PROBE EXPOSED TO VARIOUS LABORATORY SIMULATED ENVIRONMENTS.

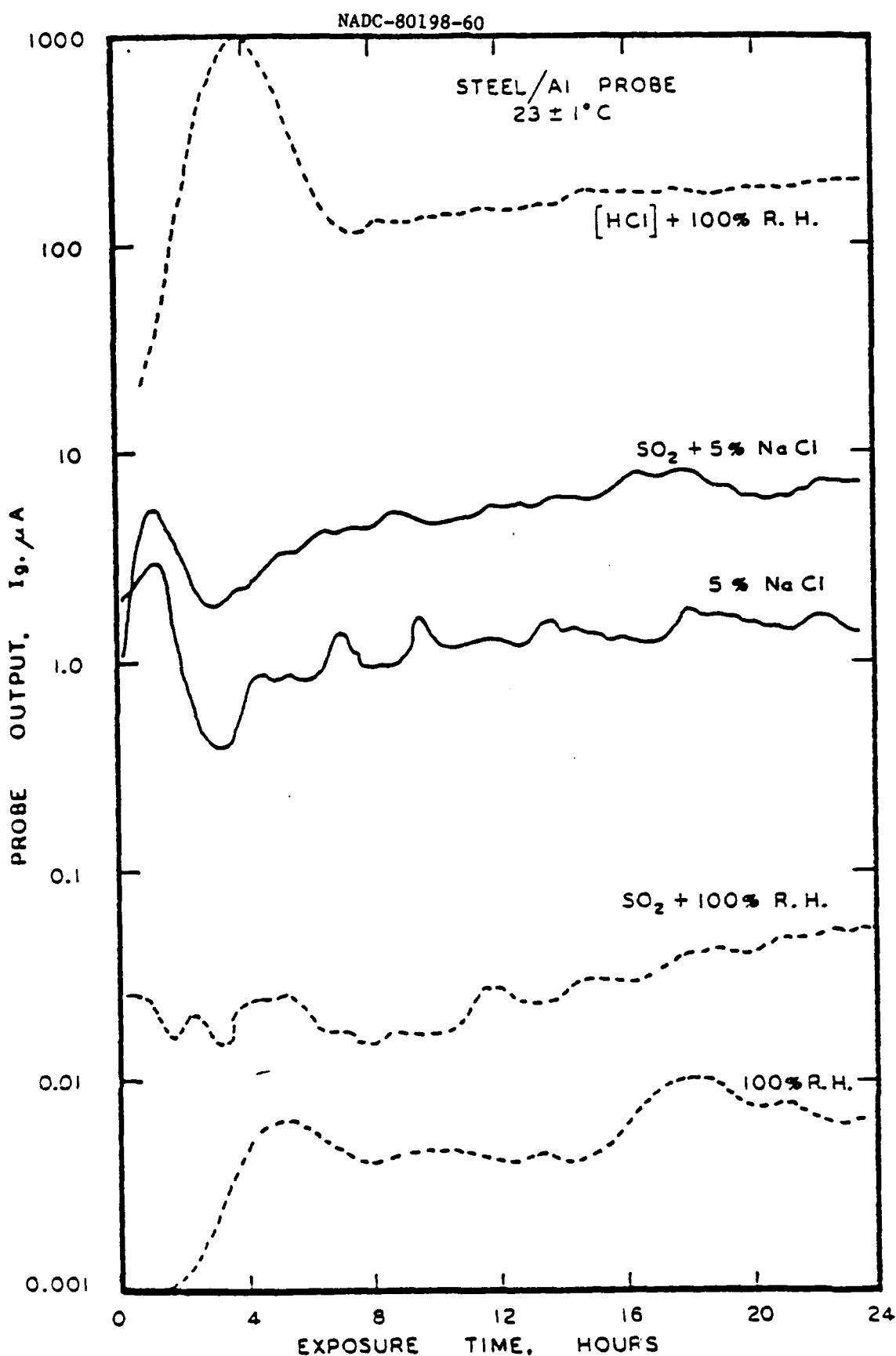


FIGURE 4. GALVANIC CURRENT TRANSIENTS OF STEEL/ALUMINUM PROBE EXPOSED TO VARIOUS LABORATORY SIMULATED ENVIRONMENTS.

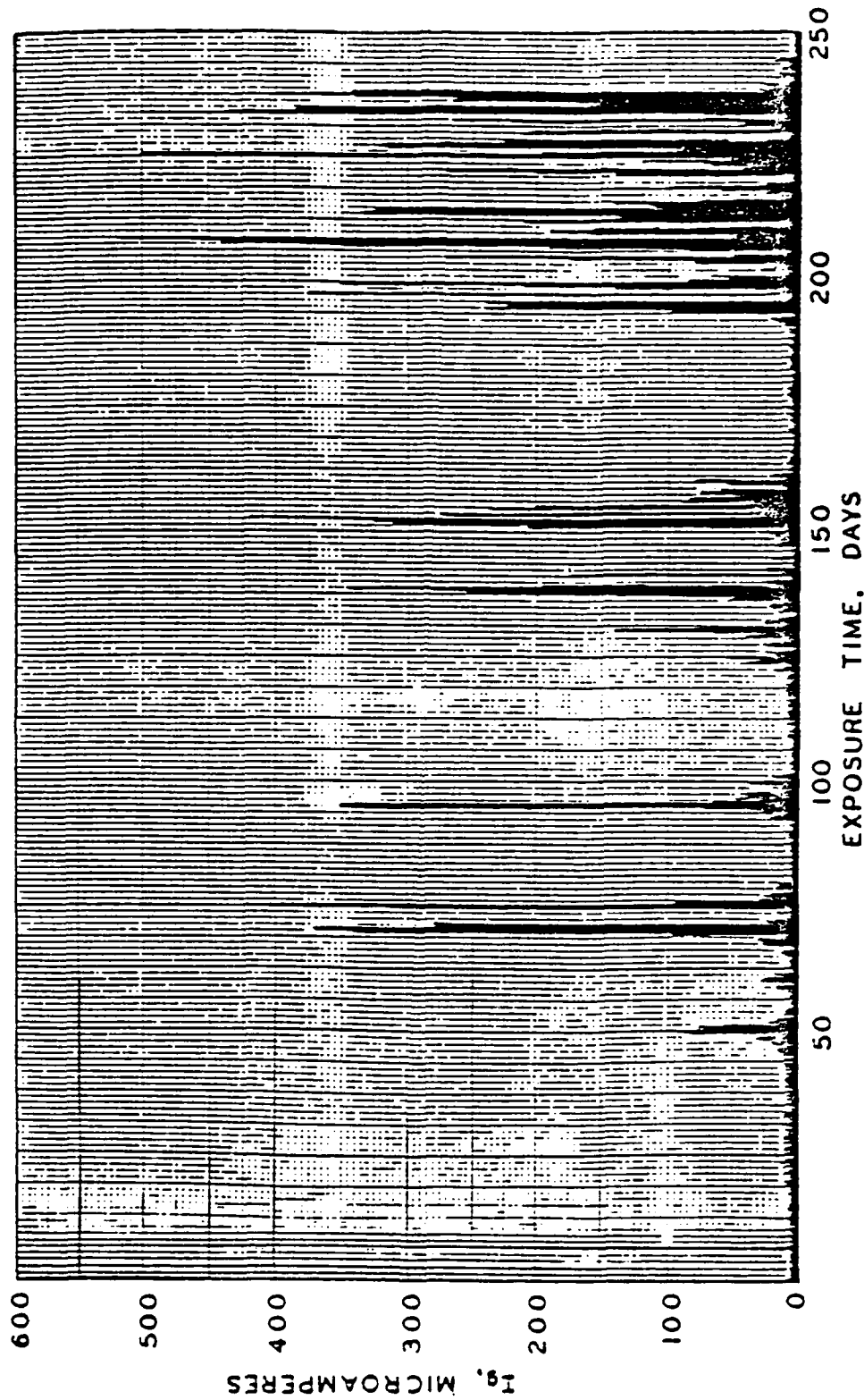


FIGURE 5. GALVANIC CURRENT TRANSIENTS OF STEEL/ALUMINUM PROBE EXPOSED TO AN AIRCRAFT CARRIER ENVIRONMENT.

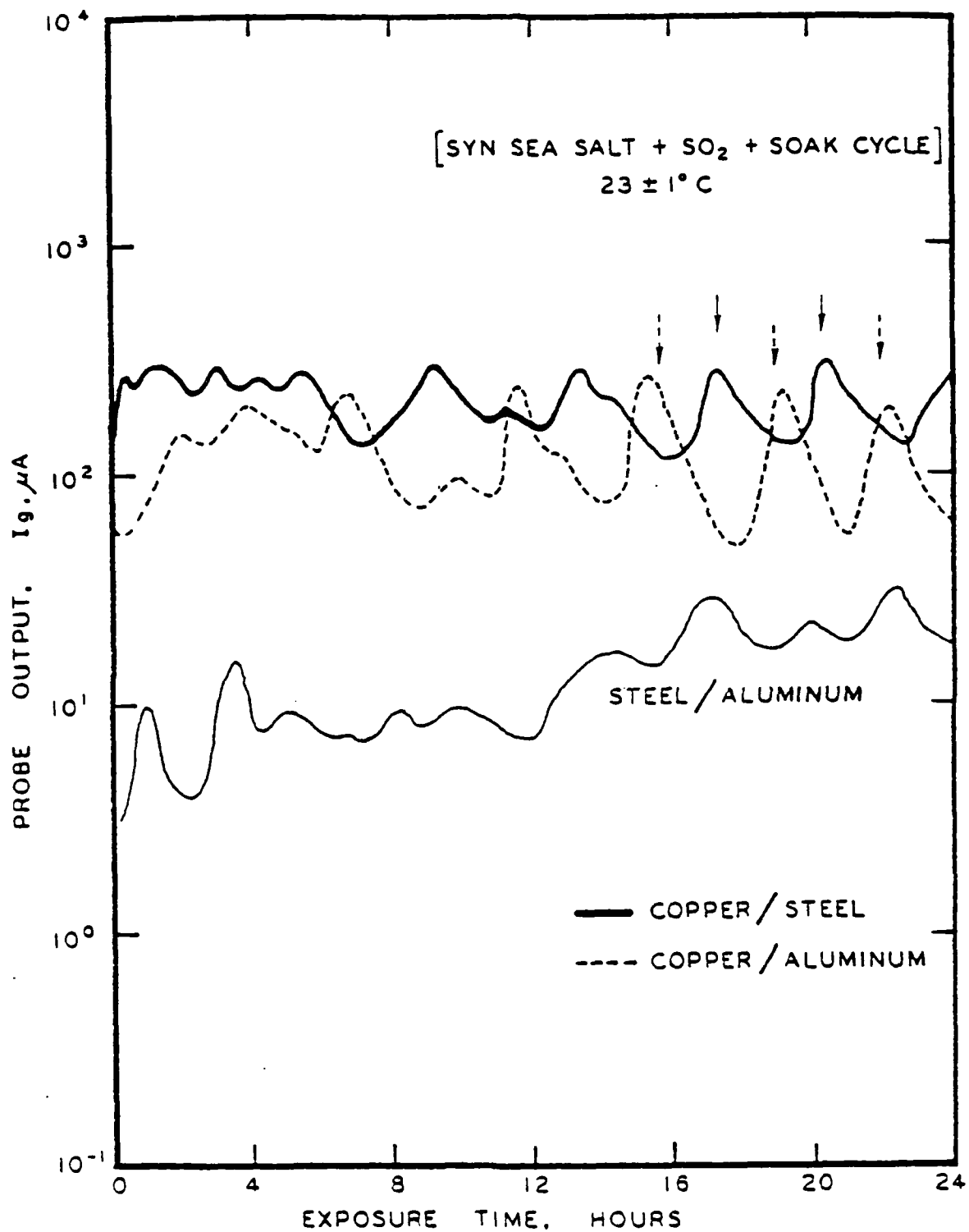


FIGURE 6. A SIMULATED CARRIER EXPOSURE TEST (SOAK CYCLE) DEMONSTRATING PROBE SENSITIVITY TO ENVIRONMENTAL CHANGES.

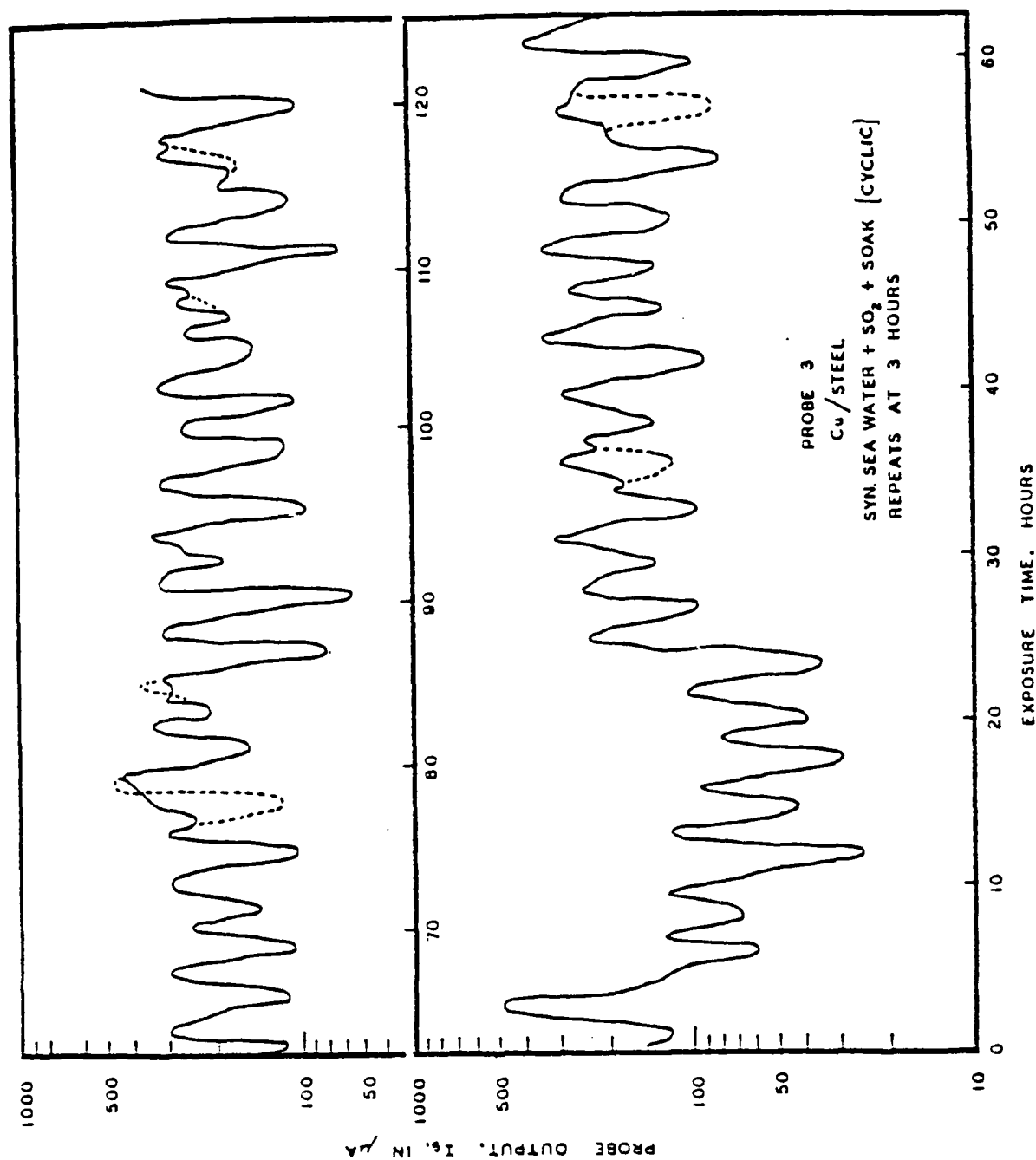


FIGURE 7. PERFORMANCE OF COPPER/STEEL PROBE IN AN ALTERNATING CORROSIVE ENVIRONMENT (SOAK CYCLE) OVER A LONG PERIOD OF EXPOSURE SHOWING THE CYCLIC NATURE OF THE ENVIRONMENTAL CORROSION TEST.

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